

Discontinuity of capacitance at the onset of surface superconductivity

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The effect of the magnetic field on a capacitor with a superconducting electrode is studied within the Ginzburg-Landau approach. It is shown that the capacitance has a discontinuity at the onset of the surface superconductivity B_{c3} which is expressed as a discontinuity in the penetration depth of the electric field into metals. Estimates show that this discontinuity is observable with recent bridges for both conventional and high- T_c superconductors of the type-II.

PACS numbers: 74.25.Op, 74.25.Nf, 85.25.-j

Capacitors based on ferroelectric layers sandwiched between metallic electrodes are approaching the technical limits of their performance. Their capacitance is not anymore exclusively given by the dielectric response of the isolating ferroelectric layer but it is reduced due to the penetration of the electric field into the metallic electrodes. Concerning the large scale integration of microscopic capacitors the penetration of the electrostatic field into the electrodes is considered as a lumped series capacitance. From the viewpoint of fundamental research, however, this phenomenon offers an opportunity to study the interaction of metallic surfaces with an applied electric field.

Here we discuss a possibility to observe the penetration of the electrostatic field into the metal in the vicinity of the transition from the normal to the superconducting state. We focus on the third critical magnetic field B_{c3} at which field the superconducting state nucleates at the surface. We predict that at this field the capacitance or the penetration of the electrostatic field possesses a jump.

The penetration of the electrostatic field into the normal metal is well understood. Ku and Ullman [1] have derived an analytic solution of the penetrating field for the jellium model within the Thomas-Fermi approximation. Their simple prediction is sufficient to explain the experimental data [2]. Much less is known about the penetration of the electrostatic field into superconductors. From the very beginning until now the history of this problem is full of contradictory concepts yielding a wide scattering of predicted values.

The question of the penetration of the electrostatic field into superconductors has been firstly addressed by the London brothers. In their early paper in 1935 they have expected that the penetration depths of the electrostatic and magnetic fields are identical [3]. One year later H. London measured the capacitance with superconducting electrodes controlled by the magnetic field and concluded that the penetration of the electrostatic field into the metal is not changed by the transition to the superconducting state [4]. While the former concept predicts

thousands of Ångströms for conventional superconductors, the latter concept suggests less than one Ångström.

Oppositely, the way from small to large penetration depths one meets in the papers by Anderson and coworkers. The Anderson theorem [5] states that the thermodynamical properties of the superconducting condensate do not depend on the electrostatic field. Accordingly, the condensate does not affect the penetration of the electrostatic field which is thus the same as in the normal metal. More recently, in the brief discussion of the effect observed by Tao, Zhang, Tang and Anderson [6], the authors speculate about a large penetration depth of the electrostatic field using ideas of the Anderson model [7] of the high- T_c superconductivity.

Apparently the problem of the electrostatic field penetrating the surface of the superconductor is far from being settled and a clear experimental message is still missing. In this letter we propose an experiment on the ferroelectric capacitor with one normal and one superconducting electrode. The magnetic field is applied to switch off the superconductivity and we will explore the vicinity of the third critical field B_{c3} .

The sensitivity of ferroelectric devices to the screening in metals is striking. Indeed, the typical Thomas-Fermi screening length in metals is about 0.5 Å, while the width L of the insulating layer has to be about a thousand of Ångströms to guarantee low leakage currents. The direct comparison of these scales is somewhat misleading, however. Taking into account the dielectric constants of the components involved we immediately obtain

$$\frac{\delta C}{C} = \frac{\epsilon_d}{\epsilon_s} \frac{\delta L}{L}. \quad (1)$$

The ceramic ferroelectric materials have $\epsilon_d \sim 10^3$ and metals have the ionic background permittivity $\epsilon_s \sim 4$ giving an enhancement factor $\epsilon_d/\epsilon_s \sim 250$. The capacitance can be measured with sensitivity better than $\delta C/C \sim 10^{-6}$, which makes it possible to observe very subtle changes of the penetration depth $\delta L \sim 10^{-5}$ Å.

First let us take a look at the interaction between the

electrostatic field and the superconductivity. The superconducting surface under the applied electrostatic field has been theoretically studied at various levels by Nabutovsky and Shapiro [8, 9, 10, 11]. They have shown that the phenomenological theory of Ginzburg and Landau (GL) yields basically the same result as the microscopic picture based on the Bogoliubov-de Gennes method. Their result was recovered in a simple form in Ref. [12], where it was shown that the effect of the applied electrostatic field E merely modifies the extrapolation length b in the de Gennes boundary condition for the GL function ψ ,

$$\frac{\nabla\psi}{\psi} = \frac{1}{b} = \frac{1}{b_0} + \frac{E}{U_s}. \quad (2)$$

This field effect is measured on the voltage scale

$$\frac{1}{U_s} = \kappa^2 \frac{\partial \ln T_c}{\partial \ln n} \frac{e^* \epsilon_s}{m^* c^2}, \quad (3)$$

where κ is the GL parameter. The logarithmic derivative of the critical temperature with respect to the electron density is of the order of unity. The need for strong applied fields follows from the ‘relativistic’ energy of an electron which is rather large, $m^* c^2 \sim 1$ MeV.

The boundary condition (2) restricts the solution of the GL equation

$$\frac{1}{2m^*} (-i\hbar\nabla - e^*\mathbf{A})^2 \psi + \alpha\psi + \beta|\psi|^2\psi = 0 \quad (4)$$

at the surface. Being non-linear, the GL equation (4) has the ability to heal any perturbation of the GL function from its optimal value on the GL coherence length $\xi = \hbar/\sqrt{2|\alpha|m^*}$. The boundary condition thus affects the GL function only in the vicinity of the surface. As consequence of that, the electric field has no remarkable effect on the bulk superconductivity.

Saint-James and de Gennes [13] have noticed that similarly to the condensation of the vapor at surfaces, the boundary condition (2) implies a nucleation of the superconducting condensate at the surface. This becomes apparent at high magnetic fields, since the bulk superconductivity vanishes at the upper critical field B_{c2} while a thin sheet of superconducting condensate survives near the surface up to fields $B_{c3} \sim 1.69461 B_{c2}$. Their result applies to the infinite extrapolation length, $1/b_0 + E/U_s = 0$.

Let us modify the method of Saint-James and de Gennes for a finite b or non-zero E . For the third critical field the GL function has an infinitesimally small amplitude so that one can neglect the cubic term in (4). As the diamagnetic current is also negligible, the vector potential reads $\mathbf{A} = (0, B_{c3}x, 0)$. We have associated the surface with the plane $x = 0$. The GL equation (4) is then solved by the parabolic cylinder function of Whit-

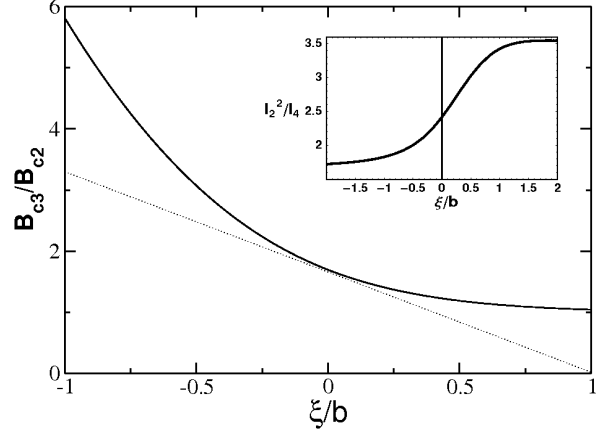


FIG. 1: The surface critical field B_{c3} versus the applied electric field E (solid line). The ratio of the third to the upper critical magnetic field relates to the argument of the parabolic cylinder function, $B_{c3}/B_{c2} = 2\nu$. The tangential line at zero bias is given as dotted line. The inset shows the field dependence of the factor in (9).

taker [14]

$$\psi(x, y, z) = N e^{iky} D_{\nu-\frac{1}{2}} \left(\frac{2x}{l} - kl \right), \quad (5)$$

with x scaled by the magnetic length $l^2 = \hbar/(eB_{c3})$ and $\nu = -\alpha m^*/\hbar e^* B_{c3} = l^2/(2\xi)^2$.

So far ν and k are parameters of the nucleating GL function ψ . We are looking for the solution with the lowest $\nu = \nu_{\min}$, because it corresponds to the highest magnetic field B_{c3} for which the nucleation is possible at fixed temperature. This highest magnetic field B_{c3} is the third critical field and its resulting value is shown in figure 1.

The dependence of the third critical field B_{c3} on the applied electrostatic field E indicates that the electrostatic field affects the surface superconductivity. The same interaction manifests itself in the effect of the magnetic field on the capacitance. The capacitance of the capacitor with one superconducting electrode reads

$$\frac{1}{C_s} = \frac{1}{C_n} - \frac{1}{\epsilon_0^2 \epsilon_s^2 S^2} \frac{\partial^2 F}{\partial E^2}, \quad (6)$$

where S is the area of the capacitor, C_n is the capacitance when both electrodes are normal, and

$$F = S \int_0^\infty dx \left[\frac{1}{2m^*} |(i\hbar\nabla + e^*\mathbf{A})\psi|^2 + \alpha|\psi|^2 + \frac{\beta}{2}|\psi|^4 \right] \quad (7)$$

is the GL free energy describing the difference between the normal and the superconducting states in the superconducting electrode.

Near the transition line $B \sim B_{c3}$, the GL function has the shape of the nucleation function (5) with $\nu = \nu_{\min}(E, B)$ and $k = k_{\min}(E, B)$. If we keep the amplitude N as a variational parameter, the free energy is its biquadratic function, $F_N = S(\alpha - \alpha_E)lN^2I_2/2 + SlN^4I_4/(4\beta)$. Here $\alpha_E = -(\hbar e^*B/m^*)\nu_{\min}(E, B)$ stands for the kinetic energy obtained from the GL function (5), while $\alpha = \alpha'(T - T_c)$ is the temperature dependent GL parameter. Integrals over powers of the parabolic cylinder functions are denoted as $I_n = \int_{k_E}^{\infty} d\tau D_{\nu_E}^n$. The condition of minimum, $\partial F_N/\partial N = 0$, is satisfied by $N_{\min}^2 = (\alpha_E - \alpha)I_2/(\beta I_4)$. From $F = F_{N_{\min}}$ or directly from (7) one obtains the free energy $F = -S(\alpha_E - \alpha)^2I_2^2/(4\beta I_4)$.

Now we can evaluate the jump of the capacitance, which appears as the magnetic field B exceeds the critical value B_{c3} . Since $\alpha_E \rightarrow \alpha$ for $B \rightarrow B_{c3}$, the discontinuity of the inverse capacitance equals

$$\frac{1}{C_s} - \frac{1}{C_n} = \frac{\hbar^2 e^* B_{c3}^2 l I_2^2}{2\epsilon_0^2 \epsilon_s m^* S \beta I_4} \left(\frac{\partial \nu_{\min}}{\partial E} \right)^2, \quad (8)$$

where we have used $\partial \alpha_E / \partial E = -(\hbar e^* B / m^*) (\partial \nu_{\min} / \partial E)$. The tangential line plotted in Fig. 1 yields $\partial \nu_{\min} / \partial E = -0.82 \xi / U_s$. The discontinuity in the capacitance is transparently expressed via the discontinuity in the penetration depth of the electric field

$$\delta L = \epsilon_0 \epsilon_s S \left(\frac{1}{C_s} - \frac{1}{C_n} \right) = \frac{0.397 \hbar^4 I_2^2}{\epsilon_0 \epsilon_s m^* \beta U_s^2 l I_4}. \quad (9)$$

In the rearrangement we have used $4\xi^2 = l^2/\nu$ and $e^* B_{c3} = 2\hbar/l^2$. From $2\nu = 1.694$ follows the numerical factor $0.82^2/(2\nu) = 0.397$. The field dependence of the factor I_2^2/I_4 is plotted as an inset in figure 1.

To further simplify expression (9) we employ the parameter $\beta = 6\pi^2 k_B^2 T_c^2 / (7\zeta(3) E_F n)$ derived from the BCS theory by Gor'kov [15], and rewrite it in terms of the BCS coherence length $\xi_{\text{BCS}} = \hbar v_F / (1.76\pi k_B T_c)$. Moreover we substitute U_s from (3) so that we obtain finally

$$\delta L = 1.86 \cdot 10^{-8} \frac{\kappa^4 \epsilon_s}{m_s^3} a_B^3 n \left(\frac{\partial \ln T_c}{\partial \ln n} \right)^2 \frac{\xi_{\text{BCS}}^2}{l}. \quad (10)$$

We have collected all universal physical constants into the Bohr radius $a_B = 4\pi\hbar^2\epsilon_0/(m_0 e^2) = 0.53 \text{ \AA}$ and the constant of fine structure $e^2/(4\pi\epsilon_0\hbar c) = 1/137$. The later appears in the fourth power giving the very small factors $1/137^4 = 2.8 \times 10^{-9}$. The mass of the Cooper pair is twice the effective mass of electrons in the metal $m^* = 2m_s m_0$ and $e^* = 2e$. For $1/b_0 + E/U_s = 0$ the factor given by the profile of the GL function is $I_2^2/I_4 = 2.42$, see figure 1.

Equation (10) is the main result of the paper. It expresses the jump in the capacitance (9) in terms of materials parameters like the logarithmic density derivative of the critical temperature, the coherence length ξ_{BCS} and the GL parameter κ . This result provides a convenient

tool to access these parameters by measuring the jump in the capacitance at the third critical field B_{c3} . Indeed, the discontinuity is small but nevertheless observable.

For an estimate we assume some typical numbers. The most sensitive measurements of capacitance performed in the $C \sim \mu\text{F}$ range are capable to monitor changes $\delta C/C \sim 10^{-6}$ with error bars at $\delta C/C \sim 10^{-7}$. From the capacitance $C = \epsilon_0 \epsilon_d S/L$ one sees that a 1000 \AA -thick dielectric layer with $\epsilon_d = 10^3$ has an optimal area of 10 mm^2 which is about the usual size of such samples [16]. The penetration depth (9) yields the relative change of the capacitance according to (1). With $\epsilon_s = 4$ and the above assumed values for the capacitance one finds that changes $|\delta L| > 3 \times 10^{-6} \text{ \AA}$ are conveniently detectable with error bars of $\delta L \sim 3 \times 10^{-7} \text{ \AA}$.

It should be noted here that these estimates remain essentially valid even if the Wagner polarization diminishing effective permittivity of thin dielectric layers is taken into account. The expected reduction of the numbers above corresponds only to a factor of ~ 2 , see Ref. [17].

Now we will show that for niobium the discontinuity falls in the range of the error bars. For niobium at temperature $T \sim 1 \text{ K}$ one can take $\kappa \sim 1.5$, see [18], and $m_s = 1.2$ giving $\kappa^4 \epsilon_s / m_s^3 = 11.7$. The logarithmic derivative is estimated in [19] as $\partial \ln T_c / \partial \ln n = 0.74$. The electron density $n = 2.2 \times 10^{28} / \text{m}^3$ yields $a_B^3 n = 3.3 \times 10^{-3}$ and the Fermi velocity $v_F = \hbar(3\pi^2 n)^{1/3} / (m_0 m_s) = 7.2 \times 10^5 \text{ m/s}$. The critical temperature $T_c = 9.5 \text{ K}$ corresponds to the BCS coherence length of $\xi_{\text{BCS}} = 3120 \text{ \AA}$. Finally we need the third critical magnetic field B_{c3} to estimate the magnetic length l . From $B_{c3} = 1.69 B_{c2}$ and the experimental value $B_{c2} = 0.35 \text{ T}$ [18] one finds $B_{c3} = 0.59 \text{ T}$, which yields $l = 325 \text{ \AA}$. With all these values we obtain from equation (10) the discontinuity $\delta L \sim 1.2 \times 10^{-6} \text{ \AA}$, which is comparable to the error bar.

There are a number of alloys [20] with the help of which one easily reaches a region of observable discontinuities. For example, 50% of niobium with 50% of tantalum has the critical temperature $T_c = 6.25 \text{ K}$ while the GL parameter at T_c is $\kappa = 3.9$ [21]. The upper critical magnetic field at $T \ll T_c$ is 0.7 T [21] giving $B_{c3} = 1.2 \text{ T}$ which yields $l = 228 \text{ \AA}$. We assume that the effective mass scales with the GL parameter so that κ/m_s remains the same as in pure niobium along with the remaining parameters. In this case, the discontinuity increases to $\delta L = 1.1 \times 10^{-5} \text{ \AA}$, which is still well observable.

We note that among intermetallic alloys there are even more promising candidates. The alloy of Nb-61% Ti has $T_c = 8.95 \text{ K}$ and $\kappa = 38.4$. The upper critical magnetic field $B_{c2} = 47 \text{ T}$ corresponds to $B_{c3} = 80 \text{ T}$, which is too high to be applied during slow measurements of the capacitance. One has to increase the temperature for the measurement so that the third critical field becomes comparable to a convenient field of 10 T , which corresponds to $l = 79 \text{ \AA}$. This estimate suggests $\delta L = 1.5 \times 10^{-4} \text{ \AA}$ which is fifty times larger than the experimental sensi-

tivity.

Detectable amplitudes of the discontinuity result also for high- T_c materials. Using the values of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ which are $T_c = 90\text{ K}$, $\kappa = 55$, $m_s = 6.92$, $\epsilon_s = 4$, $n = 5 \times 10^{27}/\text{m}^3$ [22] and $\partial \ln T_c / \partial \ln n = -2.4$ [23] as well as $l = 79\text{ \AA}$ for 10 T of the applied magnetic field, one can expect a discontinuity $\delta L = 1.4 \times 10^{-5}\text{ \AA}$.

It should be noted that the field effect on the high- T_c materials has been extensively studied within the effort to develop superconducting devices analogous to the field-effect transistors [24, 25]. There were many measurements of the field effect detecting directly changes in T_c with the applied electric field. These experiments employ the largest accessible fields because the changes in T_c are very small. The discontinuity of the capacitance can supply the missing knowledge of the field effect for low applied fields. Since the mechanism of the field effect on the high- T_c materials is not yet fully clarified, the low-field effect is of interest.

In summary, we have shown that the capacitance of the planar capacitor with one normal electrode and the other electrode to be superconducting possesses a discontinuity at the third critical field B_{c3} . This discontinuity is large enough to be observed in capacitors with ferroelectric dielectric layers of a width of 1000 \AA . We would like to point out that compared to other regions of the magneto-capacitance, the discontinuity has the advantage of being a unique feature which is not obscured by other properties of the insulator. Indeed, exploring strong electric fields one has to face the fact that the dielectric response of the ferroelectric material is non-linear. Scanning through temperatures one observes namely the Curie law of the ferroelectric transition. Moreover, the dielectric function of the ferroelectric isolator depends on the magnetic field. The measurement of the discontinuity circumvents all these problems, because all the troublesome dependencies are continuous at the onset of the surface superconductivity.

This work was supported by research plans MSM 0021620834 and No. AVOZ10100521, by grants GAČR 202/07/0597 and 202/06/0040 and GAAV 100100712 and IAA1010404, by PPP project of DAAD, by DFG Priority Program 1157 via GE1202/06 and the BMBF and by European ESF program NES.

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